

## **APPENDIX B**

# **ANALYSIS OF THE CHEMICAL FORM OF PLUTONIUM IN THE ENVIRONMENT**

### **Introduction**

In order to calculate the radiation dose to an individual from plutonium in the environment, a radiation dose analysis needs to be performed. An important part of this dose analysis is selecting the inhalation and ingestion dose conversion factors that are appropriate to the situation. In the case of plutonium, there are a number of different dose conversion factors available based on the chemical form of the plutonium being assessed. The following analysis will show that plutonium in surface soils is in the oxide form. The appropriate inhalation dose conversion factor for use in surface soils will therefore be based on the Class Y form of plutonium. An  $f_1$  value of  $10^{-05}$  will be appropriate for selecting the ingestion dose conversion factor due to the oxide form of plutonium being present.

### **Plutonium Dosimetry**

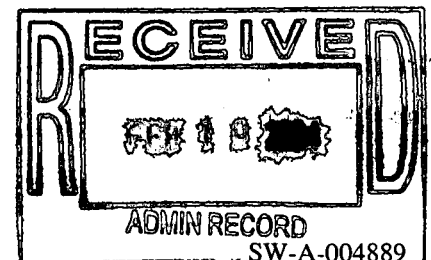
The radiation dose received by a person that inhales and/or ingests radioactive material is based on a number of biological factors. These factors include the way radioactive material moves in the lungs and the Gastrointestinal (GI) tract as well as within certain organs and tissues. This movement is examined for each radionuclide so that radionuclide specific Dose Conversion Factors (DCF) can be developed. DCFs are developed so that the radiation dose from a specific type of radioactive material can be related to a radiation dose.

Plutonium deposited in the lung is assumed to be from inhaling suspended plutonium particles. To assess this inhalation exposure, the ICRP Task Group Lung Model is used (ICRP, 1986). This lung model divides the respiratory system into four regions: the nasal passage, the tracheo-bronchiolar region, the pulmonary region and the lung associated lymph nodes. Each region is further subdivided into compartments that correspond to clearance routes from the lung. Radioactive material is

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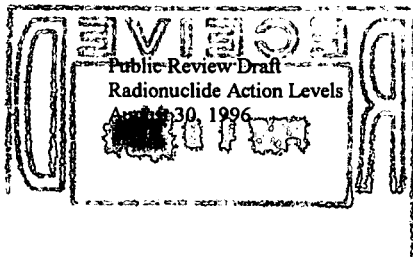
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eliminated from the lungs through clearance routes such as translocation to the blood. Three classes of compounds are broadly defined for all radioactive material deposited in the lung based on the length of time the material is retained in the lung: Class D (Clearance half-time of <10 days), Class W (Clearance half-time of 10-100 days) and Class Y (Clearance half-time of >100 days). The DCF for a radioactive material is dependent on its Class D, W or Y designation. The designation of the correct inhalation class is dependent on the chemical form of the radioactive material (i.e., oxides, nitrates, etc.). Therefore, the proper inhalation DCF can be chosen if the chemical form of a radioactive material is known. When classifying plutonium, there are two chemical forms that are important: oxides and all other forms. Oxides of plutonium are designated as being Class Y. All other chemical forms of plutonium are designated as being Class W. There are no chemical forms of plutonium associated with Class D.

Radioactive material can also enter the body through the soil ingestion and food ingestion exposure pathways. Once ingested, radioactive material is transferred to the GI tract where a fraction of the radioactive material will pass through the cells of the mucosa into the blood stream (ICRP, 1986). This fraction of the radioactive material which is transferred to the blood is called the "Fractional Absorption" or  $f_1$ . The designation of the correct ingestion  $f_1$  value is dependent on the chemical form of the radioactive material (i.e., oxides, nitrates, etc.). Therefore, the proper ingestion DCF can be chosen if the chemical form of a radioactive material is known. There are three  $f_1$  values for plutonium which relate to the chemical forms of oxides, nitrates and all other forms. These chemical forms are associated with an  $f_1$  value of  $10^{-5}$ ,  $10^{-4}$  and  $10^{-3}$ , respectively.

The magnitude of the DCF is based on the destination and fate of the radioactive material after it enters the blood. Radioactive material in the blood deposits in certain organs preferentially or is excreted. For those radioactive materials deposited in organs, the magnitude of the DCF is based on the amount of radiation absorbed by the organ. For plutonium reaching the bloodstream, 50% deposits in the skeleton, and 30% deposits in the liver (ICRP, 1986). The remaining 20% of plutonium is excreted or deposited in a variety of other organs and tissues. The plutonium is retained



in the skeleton with a retention half-time of 50 years. For the liver, the retention half-time is 20 years (ICRP, 1986).

With the above information, inhalation DCFs have been developed for Class W and Class Y plutonium separately. For the ingestion of plutonium, DCFs have been developed for each of the three potential fractional absorptions. All of these DCFs can be found in the Environmental Protection Agency's Federal Guidance Report #11 (EPA, 1988).

#### Chemical Form of Plutonium in the Environment

The chemical form of plutonium in the environment must now be selected so that an intake of plutonium can be related to a radiation dose through a DCF. The oxide, nitrate and all other chemical forms are the three potential chemical forms of plutonium that need to be assessed.

To understand which chemical form is the most applicable, the physical form of the plutonium must first be examined. In general, plutonium present in surface soils at the Rocky Flats Environmental Technology Site (RFETS) resembles the plutonium originating from the 903 Pad and Lip area. In these areas, drums containing plutonium contaminated solvents leaked to the surrounding soil. Drums were originally placed at the 903 Pad area starting in 1958 and were all removed from the area by 1968. After the drums had leaked, contaminated soils were redistributed in the area by wind and rain action (DOE, 1995). It is assumed that the plutonium in these areas are in the particulate form. A study of plutonium particulate size distributions in the eastern buffer zone showed that plutonium was in the particulate form and had a mean particle size of 0.08 micron (Rockwell, 1975). This same report stated that the particles of plutonium in the 903 Pad area had a mean particle size of 0.3 micron with maximum particle sizes of 2 to 3 microns.

The solvents in the soil at the 903 Pad and Lip either volatilized or leached into the soil fairly quickly. The plutonium particles have therefore been open to the air for about the last 30 to 35 years.

Plutonium is known to convert to plutonium dioxide when exposed to air (ANS, 1980). The rate at which oxidation occurs is based on the temperature and relative humidity of the air. At 25°C and 40% relative humidity, a 0.3 micron particle would be oxidized to plutonium dioxide in about 900 hours or 38 days (ANS, 1980). Since plutonium in the environment at RFETS has been present for a number of years, plutonium in surface soils at RFETS should be in the plutonium dioxide form.

The Operable Unit (OU) 2 Remedial Investigation (RI) Report, human health risk assessment states that plutonium in most natural environments is stable in two oxidation states, Pu III and Pu IV. In acidic environments, Pu III is the dominant species, whereas under alkaline or oxidizing conditions the dominant species is Pu IV which is solid plutonium dioxide. Since the soils at OU 2 are alkaline and oxidizing, solid plutonium dioxide should be present (DOE, 1995).

Additional studies at RFETS have demonstrated low extraction of plutonium from soil, suggesting the presence of an insoluble species (EPA, 1990). EPA states that "analyses of soil samples from Rocky Flats have shown the presence of discrete particles of plutonium (probably the oxide) attached to larger soil particles." EPA further states that, because of the stability of Pu IV relative to other forms of plutonium "most if not all plutonium in environmental...systems is in the Pu IV state."

Based on this data, the working group concluded that the plutonium present in surface soils is in the oxide form. Therefore the inhalation DCF for plutonium will be the Class Y form, and the ingestion DCF will be based on an  $f_1$  factor of  $10^{-05}$ .

## References

ANS, 1980 - Plutonium Handbook, A Guide to the Technology, Volumes I & II, American Nuclear Society, 1980

DOE, 1995 - Phase II RFI/RI Report, 903 Pad, Mound and East Trenches Area, Operable Unit Number 2, Draft Final, US Department of Energy, May 1995

EPA, 1988 - Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion and Ingestion, Federal Guidance Report #11, EPA-520/1-88-020, September 1988

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ICRP, 1986 - International Commission on Radiological Protection (ICRP) Publication 48, The metabolism of Plutonium and related Elements, April 1986

Rockwell, 1975 - Particle Size Distribution of Plutonium on Soil Surface in Rocky Flats East Buffer Zone, J. Hayden, M. DeHerrera and C. Stewart, Rockwell International, September 26, 1975

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